

Large electric field effects on the resistance of $\text{La}_{0.67}\text{Ca}_{0.33}\text{MnO}_3$ microstructures

C. Beekman, I. Komissarov and J. Aarts

Kamerlingh Onnes Laboratorium, Leiden University, The Netherlands*

(Dated: January 26, 2013)

We investigate electric field effects in thin film microbridges of $\text{La}_{0.7}\text{Ca}_{0.3}\text{MnO}_3$ with the focus on the regime of metal-insulator transition. A mechanically milled SrTiO_3 substrate is used as a backgate dielectric. Inside the metal-insulator transition we find a strong unipolar field-induced reduction in resistance, as well as a suppression of the nonlinear features in the I-V curves we observed earlier. We associate the observed effects with a phase separated state in which metallic regions coexist with short range correlated polaron regions. When the glassy polaron phase has fully developed, and closes off the microbridge, the field effects disappear leaving the strongly nonlinear behavior of the transport current unaltered.

Conventional field effect transistors (FET) consist of semiconductor channels in which the carrier density is modulated by an electric field applied through a gate on top or below the channel. The dimensions of these semiconductor microstructures are reaching their intrinsic physical limit. Further increase of circuit density requires the consideration of different materials for both the gate dielectric and the channel. Promising are correlated electron systems based on Mott insulators, such as high T_C superconductors[1] and Colossal Magnetoresistance (CMR) manganites [2]. The abundant amount of potential carriers (d-electron) and the ability to control the bandgap could result in novel FET-type devices.

Doped manganites such as $\text{La}_{0.7}\text{Ca}_{0.3}\text{MnO}_3$ (L7CMO), obtained by hole doping the antiferromagnetic insulating parent compound LaMnO_3 , show a large variety in physical properties [3, 4]. The Ca-doping introduces mixed Mn-valence, with both Mn^{3+} present, which is Jahn-Teller (JT) active, and Mn^{4+} , which is not. This leads to competing interactions, trapping of electrons in JT distortions (polarons) and itinerancy of the electrons in the Double Exchange (DE) mechanism[5] when spins become polarized. Depending on the doping, this leads to a metal-to-insulator transition (from low to high temperature), at a characteristic temperature T_{MI} . Epitaxial all perovskite FET-devices already have been under investigation. For example a 3 nm $\text{La}_{0.8}\text{Sr}_{0.2}\text{MnO}_3$ film (screening length less than 1 nm[6],) combined with a ferroelectric material ($\text{PbZr}_{0.2}\text{Ti}_{0.8}\text{O}_3$, PZT) as the gate dielectric, shows E -field induced modulation of T_{MI} and the magnetoresistance. These effects [6, 7] are *bipolar* (i.e. the sign of the resistance change is opposite for opposite signs of the applied gate voltage) and attributed to the modulation of the charge carrier density in the manganite, in other words a straightforward modulation of the doping effect. In a different investigation, large in-plane resistance variations ($\sim 76\%$, bipolar) were observed in a 50 nm thick L7CMO films[8] with the PZT-gate geometry, and smaller effects for devices deposited on an SrTiO_3 (STO) film as the backgate dielectric. Since the thickness of the film is much larger than the E -field screening length, the effects were attributed to the presence of a phase separated state, the more so since there was clear asymmetry for the two signs of the gate voltage. For phase

separation, fully *unipolar* field effects were also observed, for example in $\text{La}_{0.8}\text{Ca}_{0.2}\text{MnO}_3$ thin films on mechanically thinned STO substrates [9].

What has been little investigated is the effect of an applied E -field on a microbridge, where the channel width may be comparable to the length scales associated with the phase separation phenomenon. Here we report E -field effects on strained L7CMO microbridges using the STO substrate as a backgate dielectric. Our microbridges show a metal-insulator transition at T_{MI} and the well-known CMR effect, typical for strained thin films. They also show non-linear current I - voltage V characteristics in the range of the MI-transition, on which we reported before [10]. We find strong *unipolar* field effects in the onset of the transition, which we associate with the occurrence of a phase separated state, in which metallic regions coexist with short-range correlated polaron regions[11]. As the system is warmed through the M-I transition the field effects disappear when the more or less homogeneous correlated polaron (glass) phase is fully developed.

The L7CMO films were grown by DC-sputtering in an oxygen atmosphere of 3 mbar and at a growth temperature of 840°C . The films are patterned into microbridges[12] using electron-beam lithography and Ar-etching (see Fig.1b). The STO substrate was subsequently mechanically milled down to $100\text{ }\mu\text{m}$ and used as a gate dielectric. Measurements before

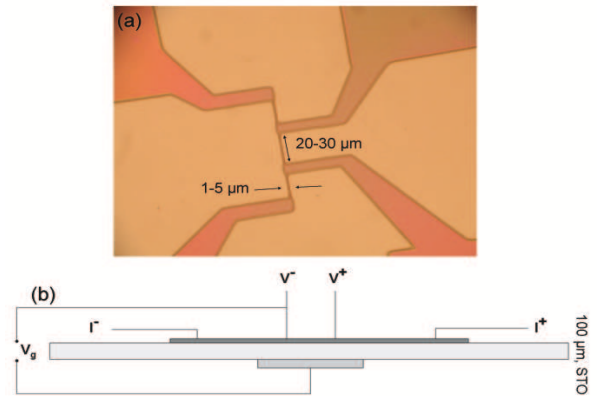


FIG. 1: a) The microbridge patterned into the L7CMO thin film. Dimensions: width: $5\text{ }\mu\text{m}$ and the distance between the voltage contacts is $30\text{ }\mu\text{m}$. b) FET device geometry.

*E-mail: aarts@physics.leidenuniv.nl

FIG. 2: $R(T)$ behavior (left axis) of a $500 \times 10 \text{ nm}^2$ L7CMO bridge determined from I-V curves at I around $0.1 \mu\text{A}$ for three applied gate voltages: $V_g = 0, \pm 75 \text{ V}$. On the right axis the ratio between $R(0 \text{ V})$ and $R(-75 \text{ V})$ is shown.

and after the milling show that the bridges remain undamaged during this process. The geometry of the measured devices is shown in Fig.1a. The gate voltage V_g is applied between the back of the STO substrate (through a silver paint contact) and one of the voltage contacts of the microbridge. We measured I-V curves as function of temperature and in high magnetic fields using a Physical Properties Measurement System (Quantum Design) for temperature and magnetic field control ($T = 20 - 300 \text{ K}$; $H_a = 0 - 9 \text{ T}$). We found the leak currents through the gate to be negligible compared to the currents used for the I-V measurements.

I-V characteristics were measured in applied electric fields up to $1 \times 10^6 \text{ V/m}$ for microbridges patterned in 10 nm thick L7CMO films grown on STO substrates. In Fig.2 we show the resistance behavior of the $0.5 \mu\text{m}$ bridge in zero field and upon application of $\pm 75 \text{ V}$ (i.e. $7.5 \times 10^5 \text{ V/m}$). In zero field we find typical resistance vs. temperature behavior for strained L7CMO films[13]. Furthermore, we observe a strong E -field effect in the transition around $T = 120 \text{ K}$ ($T_{MI} \approx 160 \text{ K}$). The resistance is reduced by a factor of 2 when $V_g = -75 \text{ V}$ is applied. Application of a positive gate voltage shows a smaller reduction, the effect is *unipolar*. It is important to note is that the position of T_{MI} remains unchanged when gate voltages are applied, and also that the effect is sharply peaking in the regime of the MI transition and not beyond, quite different from the observations by Wu *et al.*[8] Repeating this measurement on a $5 \mu\text{m}$ bridge (Fig.3) we observe a similar effect. In this case the effect is also sharply peaked, now at $T = 90 \text{ K}$ ($T_{MI} \approx 120 \text{ K}$), and the resistance is reduced by a factor of 5 upon application of $V_g = +75 \text{ V}$, with a somewhat larger asymmetry (compared to the $0.5 \mu\text{m}$ microbridge) between gate voltages of opposite sign. In both microbridges we also observe a (smaller) reduction in resistance at low temperatures. Both signs of the gate voltage result in similar resistance changes. In this temperature regime, once an E -field has been applied the microbridge appears to be irreversibly changed. For the microbridge in Fig.3 the initial low temperature (higher resistivity) state was not recovered after gating. However, repetition of the same experiment but with application of 100 V did show very similar behavior around the transition albeit with slightly reduced magnitude for the field induced resistance drop (data not shown).

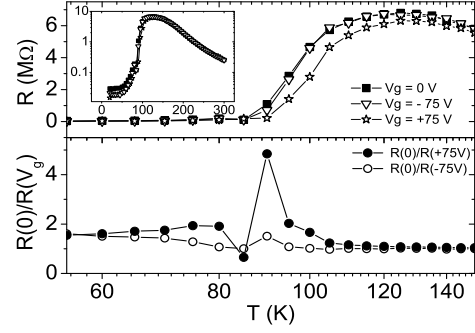


FIG. 3: (Top) $R(T)$ behavior of a $5 \mu\text{m} \times 10 \text{ nm}$ L7CMO bridge measured at $I = 0.1 \mu\text{A}$ for three applied gate voltages: $V_g = 0, \pm 75 \text{ V}$ (Inset: full R vs. T curves). (Bottom) ratio between $R(0)$ and $R(V_g)$ (closed circles: $V_g = +75 \text{ V}$; open circles: $V_g = -75 \text{ V}$).

Next we turn to the I-V curves measured on the $5 \mu\text{m}$ sample. In accordance with previous findings[10] they are linear for most temperatures but show strong nonlinear behavior in the steep part of the transition. Here we investigate the influence of the applied electric field on these nonlinearities. Fig.4 shows two I-V curves ($5 \mu\text{m}$ bridge, $T = 100 \text{ K}$ and $T = 110 \text{ K}$) and their corresponding (numerical) derivatives for $V_g = 0 \text{ V}$ and $V_g = +100 \text{ V}$. At 100 K the nonlinearities which we associated with the formation of a homogeneous glassy polaron phase start to appear, with a full width of the peak in dV/dI of $1 \mu\text{A}$. From the data it becomes clear that at this temperature the nonlinearity is suppressed upon application of an E -field. At $T = 110 \text{ K}$, where the nonlinearity has developed strongly (peak width: $4 \mu\text{A}$), the strength and shape of the peak remains fully unaltered when an E -field is applied.

One concern with respect to our observations might be that they are connected the cubic-to-tetragonal phase transition in STO around 105 K . A small effect was actually found to exist in thin films of L7CMO on STO, not in the resistivity, but in the temperature coefficient (TC) $1/\rho(d\rho/dT)$ (with ρ the specific resistance), which showed a variation of 0.5% in a 9 nm

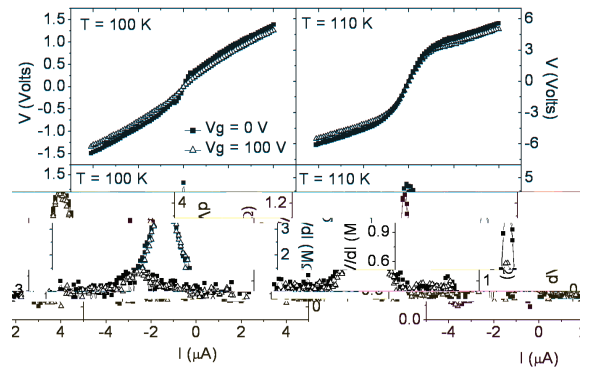


FIG. 4: (Top) I-V curves for $5 \mu\text{m}$ bridge at $T = 100 \text{ K}$ and $T = 110 \text{ K}$. (Bottom) corresponding numerical derivatives. $V_g = 0 \text{ V}$ (closed squares) and $V_g = +100 \text{ V}$ (open triangles).

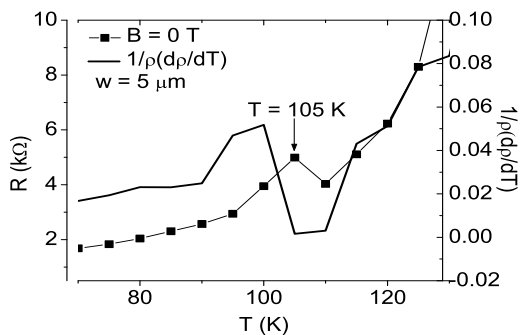


FIG. 5: $R(T)$ behavior of a 20 nm thick L7CMO microbridge ($w = 5 \mu\text{m}$) between $T = 50 - 135$ K. Right axis: $1/\rho(d\rho/dT)$ (solid line). The arrow indicates the transition in the STO. Inset: full $R(T)$ curve.

film[15]. In that film T_p was at 160 K, and the TC variation was observed in the flat metallic part of the resistance. Our thin films have lower T_p , which we have argued is the effect of homogeneous strain[13], and a 1% variation is not visible in the strong decrease in ρ below the MI transition. We could detect a variation in the TC in the microbridge of a 20 nm film where $T_p \approx 160$ K. As shown in Fig.5 we even find a variation in $R(T)$, which was not yet reported before. This is clearly due to the larger film homogeneity in the small structure. Another point to be made is that there is hardly any change in the dielectric constant of STO at the phase transition[16]. Our E -field effects appear to be intrinsic features of the L7CMO microstructures.

We explain the observations in light of our previous report on the formation of a polaron glass phase in L7CMO microbridges as it is warmed through the M-I transition[10]. The *unipolar* nature of the observed effects indicate that they are to be attributed to a state in which metallic and insulating re-

gions coexist, and are influenced by the E -field. The insulating regions are formed by short range correlated polarons, as precursor to the polaronic state at high temperature. The applied field changes the relative volume fraction of the coexisting phases by accumulating charge at the interfaces between them, which can result in the dielectric breakdown of the inhomogeneities. Important to note is that the maximum in the effect occurs in the onset of the transition when the nonlinearities just start to appear in the I - V curves. When the nonlinear effect has fully developed, the electric field effect disappears. Apparently, when the glassy polaron phase becomes homogeneous and closes off the bridge, the E -field cannot break it down. We note that the effect in the transition is unipolar but asymmetric. It is possible that doping still plays a role and that asymmetry in hole and electron modulation of inhomogeneities in the microbridge lead to the observed asymmetric behavior.

We also note that the effects in the transition are different from those at low temperatures, where it is irreversible or hysteretic, indicating that this regime is also not inhomogeneous, and that the collapse and rebuilding of the insulating regions is not well controlled [17]. The microbridge on the other hand appears to at least mostly relax back to its initial state since remeasuring the gate effect (days later) leads to similar but somewhat smaller effects in the transition.

In conclusion, going to micron-sized structures reveals a strong response of $\text{La}_{0.7}\text{Ca}_{0.3}\text{MnO}_3$ thin films to an applied electric field which has not been reported before, and which is clearly tied to the percolating behavior of the conductance in the bridge, which takes place on the scale of the width of the bridge.

We are grateful for discussions with J. Zaanen. This work was part of the research program of the Stichting F.O.M., which is financially supported by NWO.

-
- [1] C.H. Ahn et al., Science **284**, 1152 (1999)
 - [2] C.H. Ahn et al., Rev. Mod. Phys. **78**, 1185 (2006)
 - [3] Colossal Magnetoresistive oxides, Y. Tokura (CRC Press, Taylor & Francis Publishing group, 2000).
 - [4] Nanoscale Phase Separation and Colossal Magnetoresistance, E. Dagotto (Springer Series in Solid State Sciences volume 136, 2003).
 - [5] C. Zener, Phys. Rev. **82**, 403 (1951).
 - [6] X. Hong, A. Posadas, A. Lin, and C. H. Ahn, Phys. Rev. B, **68**, 134415 (2003)
 - [7] I. Pallechi, L. Pellegrino, E. Bellingeri, A.S. Siri, and D. Marré, Appl. Phys. Lett. **83**, 4435 (2003)
 - [8] T. Wu, S.B. Ogale, J.E. Garrison, B. Nagaraj, Amlan Biswas, Z. Chen, R.L. Greene, R. Ramesh, and T. Venkatesan, Phys. Rev. Lett. **86**, 5998 (2001).
 - [9] M. Eblen-Zayas, A. Bhattacharya, N.E. Staley, A.L. Kobrinskii, and A.M. Goldman, Phys. Rev. Lett. **94**, 037204 (2005)
 - [10] C. Beekman, J. Zaanen and J.Aarts, arXiv:1009.1386v1 [cond-mat.str-el] (accepted for publication in Phys. Rev. B).
 - [11] J.W. Lynn, D.N. Argyriou, Y. Ren, Y. Chen, Y.M. Mukovskii, and D.A. Shulyatev, Phys. Rev. B **76**, 014437 (2007)
 - [12] C. Beekman, I. Komissarov, M. Hesselberth, and J. Aarts, Appl. Phys. Lett. **91**, 062101 (2007).
 - [13] C. Beekman, M. Porcu, H.W. Zandbergen and J. Aarts, Phys. Rev B, submitted (ArXiv)
 - [14] D.C. Meyer, A.A. Levin, S. Bayer, A. Gorbunov, W. Pompe, and P. Paufler, Appl. Phys. A, **80**, 515 (2005).
 - [15] M. Egilmez, M. M. Saber, I. Fan, K. H. Chow, and J. Jung, Phys. Rev. B, **78**, 172405 (2008).
 - [16] E.K.H. Salje, B. Wruk, and S. Marais, Ferroelectrics **124**, 185 (1991).
 - [17] S. Dong, C. Zhu, Y. Wang, F. Yuan, K.F. Wang, and J.-M. Liu, J. Phys. Condens. Matt. **19**, 266202 (2007).